

## REPORT DOCUMENTATION PAGE

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14. ABSTRACT This grant enabled the exploration of 4d and 5d transition metal oxides, in particular, ruthenates, as well as novel two-dimensional (2D) electronic materials, all featuring interesting physical properties and/or structure-property relationships. Specifically, we carried out electrical and magneto transport measurements on La <sub>4</sub> Ru <sub>6</sub> O <sub>19</sub> , BaRuO <sub>3</sub> , and Sr <sub>2</sub> RuO <sub>4</sub> single crystals, and pursued the characterization, device fabrication, as well as the exploration of the potential uses of 2D electronic materials including graphene and NbSe <sub>2</sub> . La <sub>4</sub> Ru <sub>6</sub> O <sub>19</sub> showed a ferromagnetic quantum criticality near ambient pressure, four layered hexagonal (ATI) and nine layered rhombohedral (ODI)		
15. SUBJECT TERMS Quantum materials; structure-property relationship, quantum transport; low-temperature magneto transport measurements		
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## **Report Title**

Final Report: Quantum Material Properties of 4d and 5d Transition Metal Oxides and Potential Applications

### **ABSTRACT**

This grant enabled the exploration of 4d and 5d transition metal oxides, in particular, ruthenates, as well as novel two-dimensional (2D) electronic materials, all featuring interesting physical properties and/or structure-property relationships. Specifically, we carried out electrical and magneto transport measurements on La<sub>4</sub>Ru<sub>6</sub>O<sub>19</sub>, BaRuO<sub>3</sub>, and Sr<sub>2</sub>RuO<sub>4</sub> single crystals, and pursued the characterization, device fabrication, as well as the exploration of the potential uses of 2D electronic materials including graphene and NbSe<sub>2</sub>. La<sub>4</sub>Ru<sub>6</sub>O<sub>19</sub> showed a ferromagnetic quantum criticality near ambient pressure; four-layered hexagonal (4H) and nine-layered rhombohedral (9R) BaRuO<sub>3</sub> possess interesting structure-property relationships, with the strong one-dimensional character in the 9R structure leads to clear deviation from the metallic behavior seen in the 4H structure. The Nernst effect signal in the normal state of Sr<sub>2</sub>RuO<sub>4</sub> is large which decreases linearly as a function of temperature, possibly related to the band-dependent magnetic fluctuation, which was suppressed by the emergence of coherence at low temperatures. We observed strong magneto conductance fluctuations in few layer graphene near the charge neutral point, and novel electronic states at the “interface” between monolayer and bilayer graphene. For NbSe<sub>2</sub>, we found that the CDW was suppressed in the atomically thin NbSe<sub>2</sub> while SC survives down to two atomic layers.

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**Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:**

**(a) Papers published in peer-reviewed journals (N/A for none)**

Received      Paper

- 05/25/2015 1.00 X. F. Xu, Z. A. Xu, T. J. Liu, D. Fobes, Z. Q. Mao, J. L. Luo, Y. Liu. Band-Dependent Normal-State Coherence in Sr<sub>2</sub>RuO<sub>4</sub>:Evidence from Nernst Effect and Thermopower Measurements, Physical Review Letters, (07 2008): 0. doi: 10.1103/PhysRevLett.101.057002
- 05/25/2015 2.00 Joshua A. Robinson, Neal E. Staley, Conor P. Puls, Joseph P. Stitt, Mark A. Fanton, Konstantin V. Emtsev, Thomas Seyller, Ying Liu. Raman Topography and Strain Uniformity of Large-Area Epitaxial Graphene, Nano Letters, (03 2009): 0. doi: 10.1021/nl802852p
- 05/25/2015 3.00 C. P. Puls, N. E. Staley, Y. Liu. Interface states and anomalous quantum oscillations in hybrid graphene structures, Physical Review B, (06 2009): 0. doi: 10.1103/PhysRevB.79.235415
- 05/25/2015 4.00 Ying Liu, Linjun Li, Zhuan Xu, Peter Eklund, Neal E. Staley, Jian Wu. Electric field effect on superconductivity in atomically thin flakes of NbSe<sub>2</sub>, Physical Review B, (11 2009): 0. doi: 10.1103/PhysRevB.80.184505
- 05/26/2015 5.00 Y. Liu, I. G. Deac, P. Khalifah, R. J. Cava, Y. A. Ying, P. Schiffer, K. D. Nelson. Possible observation of quantum ferromagnetic fluctuations in La<sub>4</sub>Ru<sub>6</sub>O<sub>19</sub>, Physical Review B, (07 2009): 0. doi: 10.1103/PhysRevB.80.024303
- 05/26/2015 6.00 Y. A. Ying, Y. Xin, B. W. Clouser, E. Hao, N. E. Staley, Y. Liu, R. J. Myers, L. F. Allard, D. Fobes, T. Liu, Z. Q. Mao. Suppression of Proximity Effect and the Enhancement of p-Wave Superconductivityin the Sr<sub>2</sub>RuO<sub>4</sub>-Ru System, Physical Review Letters, (12 2009): 0. doi: 10.1103/PhysRevLett.103.247004
- 05/26/2015 7.00 Ying Liu. Phase-sensitive-measurement determination of odd-parity, spin-triplet superconductivity in Sr<sub>2</sub>RuO<sub>4</sub>, New Journal of Physics, (07 2010): 0. doi: 10.1088/1367-2630/12/7/075001
- 05/26/2015 8.00 Y. A. Ying, Y. Liu, T. He, R. J. Cava. Magnetotransport properties of BaRuO<sub>3</sub>: Observation of two scattering rates, Physical Review B, (12 2011): 0. doi: 10.1103/PhysRevB.84.233104

**TOTAL:**      **8**

**Number of Papers published in peer-reviewed journals:**

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**(b) Papers published in non-peer-reviewed journals (N/A for none)**

Received      Paper

**TOTAL:**

**Number of Papers published in non peer-reviewed journals:**

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**(c) Presentations**

November 30, 2007. Department of Physics, University of Illinois in Urbana Champaign. "Pairing symmetries of the bulk and the 3-K phases of Sr<sub>2</sub>RuO<sub>4</sub>."

December 11, 2007, Workshop on Physics of Sr<sub>2</sub>RuO<sub>4</sub>, Kavli Institute for Theoretical Physics, University of California Santa Barbara. "Pairing symmetry of the 3-K phases of Sr<sub>2</sub>RuO<sub>4</sub>."

January 15, 2009. Argonne National Laboratory, Chicago. "From Graphene to Single-Sheet Superconductors."

January 16, 2009. Northwestern University, Evanston. "Spin-triplet Superconductivity in Sr<sub>2</sub>RuO<sub>4</sub>."

**Number of Presentations:** 4.00

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**Non Peer-Reviewed Conference Proceeding publications (other than abstracts):**

Received      Paper

**TOTAL:**

**Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):**

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**Peer-Reviewed Conference Proceeding publications (other than abstracts):**

Received      Paper

**TOTAL:**

**Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):**

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**(d) Manuscripts**

Received      Paper

**TOTAL:**

**Number of Manuscripts:**

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**Books**

Received      Book

**TOTAL:**

Received      Book Chapter

**TOTAL:**

**Patents Submitted**

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**Patents Awarded**

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**Awards**

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### **Graduate Students**

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	Discipline
Ying, Yiqun, A	0.50	
<b>FTE Equivalent:</b>	<b>0.50</b>	
<b>Total Number:</b>	<b>1</b>	

### **Names of Post Doctorates**

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
<b>FTE Equivalent:</b>	
<b>Total Number:</b>	

### **Names of Faculty Supported**

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	National Academy Member
Liu, Ying	0.08	
<b>FTE Equivalent:</b>	<b>0.08</b>	
<b>Total Number:</b>	<b>1</b>	

### **Names of Under Graduate students supported**

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
<b>FTE Equivalent:</b>	
<b>Total Number:</b>	

### **Student Metrics**

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: ..... 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:..... 0.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):..... 0.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense ..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields: ..... 0.00

### **Names of Personnel receiving masters degrees**

<u>NAME</u>
-------------

| **Total Number:** |

## Names of personnel receiving PhDs

<u>NAME</u>
Ying, Yiqun, A
<b>Total Number:</b>

1

## Names of other research staff

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
<b>FTE Equivalent:</b>	
<b>Total Number:</b>	

## Sub Contractors (DD882)

## Inventions (DD882)

## Scientific Progress

This grant enabled the exploration of 4d and 5d transition metal oxides, in particular, ruthenates, as well as novel two-dimensional (2D) electronic materials, all featuring interesting physical properties and/or structure-property relationships. Specifically, we carried out electrical and magneto transport measurements on La<sub>4</sub>Ru<sub>6</sub>O<sub>19</sub>, BaRuO<sub>3</sub>, and Sr<sub>2</sub>RuO<sub>4</sub> single crystals, and pursued the characterization, device fabrication, as well as the exploration of the potential uses of 2D electronic materials including graphene and NbSe<sub>2</sub>.

Measurements on single crystals La<sub>4</sub>Ru<sub>6</sub>O<sub>19</sub> revealed that the previously observed metal-metal bonding and non-Fermi-liquid behavior are likely due to the existence of a ferromagnetic quantum criticality near ambient pressure in this material. Our results obtained on single crystals of four-layered hexagonal (4H) and nine-layered rhombohedral (9R) BaRuO<sub>3</sub> provided insight into the structure-property relationships of BaRuO<sub>3</sub> polymorphs, with the strong one dimensional character in the 9R structure leads to clear deviation from the metallic behavior seen in the 4H structure. Measurements on the Nernst effect in the normal state of Sr<sub>2</sub>RuO<sub>4</sub> found a large value of the Nernst signal with its magnitude increasing with the decreasing temperature until reaching a maximum around 20–25 K, below which it starts to decrease linearly as a function of temperature. The observed behavior was explained by relating the Nernst signal to the band-dependent magnetic fluctuation suppressed by the emergence of coherence in one of the bands at low temperatures in Sr<sub>2</sub>RuO<sub>4</sub>. In addition, preliminary work on ionic liquid gating of Ca<sub>3</sub>Ru<sub>2</sub>O<sub>7</sub>, the n = 2 member of the Roddelsden-Popper series of Can+1RunO<sub>3n+1</sub> aiming at tuning its structural transition by electric field was carried out. We found that the effect of electric field is minimal.

We also carried out work on graphene, which was focused on the device fabrication and characterization on graphene flakes prepared by mechanical exfoliation from bulk graphite, and NbSe<sub>2</sub>, on the interplay between charge-density waves (CDWs) and superconductivity (SC) in atomically thin flakes again prepared by mechanical exfoliation from the bulk crystals. We observed strong magneto conductance fluctuations in few layer graphene near the charge neutral point, and novel electronic states at the “interface” between monolayer and bilayer graphene. For NbSe<sub>2</sub>, we found that the CDW was suppressed easily in the atomically thin NbSe<sub>2</sub> while SC survives at least down to two atomic layers.

## Technology Transfer

**Report Type: Final Report**

**Proposal Number: 52615PH**

**Agreement Number: W911NF0710182**

**Proposal Title:** **Quantum Material Properties of 4d and 5d Transition Metal Oxides and Potential Applications**

**Report Period Begin Date:** **06/01/2007**

**Report Period End Date:** **05/31/2011**

**Program monitor:** **Dr. Marc Ulrich**

**Prepared by Ying Liu**

**Abstract**

This grant enabled the exploration of 4d and 5d transition metal oxides, in particular, ruthenates, as well as novel two-dimensional (2D) electronic materials, all featuring interesting physical properties and/or structure-property relationships. Specifically, we carried out electrical and magneto transport measurements on La<sub>4</sub>Ru<sub>6</sub>O<sub>19</sub>, BaRuO<sub>3</sub>, and Sr<sub>2</sub>RuO<sub>4</sub> single crystals, and pursued the characterization, device fabrication, as well as the exploration of the potential uses of 2D electronic materials including graphene and NbSe<sub>2</sub>. La<sub>4</sub>Ru<sub>6</sub>O<sub>19</sub> showed a ferromagnetic quantum criticality near ambient pressure; four-layered hexagonal (4H) and nine-layered rhombohedral (9R) BaRuO<sub>3</sub> possess interesting structure-property relationships, with the strong one-dimensional character in the 9R structure leads to clear deviation from the metallic behavior seen in the 4H structure. The Nernst effect signal in the normal state of Sr<sub>2</sub>RuO<sub>4</sub> is large which decreases linearly as a function of temperature, possibly related to the band-dependent magnetic fluctuation, which was suppressed by the emergence of coherence at low temperatures. We observed strong magneto conductance fluctuations in few layer graphene near the charge neutral point, and novel electronic states at the “interface” between monolayer and bilayer graphene. For NbSe<sub>2</sub>, we found that the CDW was suppressed in the atomically thin NbSe<sub>2</sub> while SC survives down to two atomic layers.

Subject terms:

Quantum materials; structure-property relationship, quantum transport; low-temperature magneto transport measurements

**Main results**

Measurements on single crystals La<sub>4</sub>Ru<sub>6</sub>O<sub>19</sub> with metal-metal bonding revealed that the previously observed non-Fermi-liquid behavior are likely due to the existence of a ferromagnetic quantum criticality near ambient pressure in this material. Our results obtained on single crystals of four-layered hexagonal (4H) and nine-layered rhombohedral (9R) BaRuO<sub>3</sub> provided insight

into the structure-property relationships of BaRuO<sub>3</sub> polymorphs, with the strong one-dimensional character in the 9R structure leads to clear deviation from the metallic behavior seen in the 4H structure. Measurements on the Nernst effect in the normal state of Sr<sub>2</sub>RuO<sub>4</sub> found a large value of the Nernst signal which decreases linearly as a function of temperature. The observed behavior was explained by relating the Nernst signal to the band-dependent magnetic fluctuation suppressed by the emergence of coherence in one of the bands at low temperatures in Sr<sub>2</sub>RuO<sub>4</sub>. We also found that the Ru/Sr<sub>2</sub>RuO<sub>4</sub> interface in the eutectic phase of Ru-Sr<sub>2</sub>RuO<sub>4</sub> is atomically sharp. In addition, preliminary work on ionic liquid gating of Ca<sub>3</sub>Ru<sub>2</sub>O<sub>7</sub>, the n = 2 member of the Roddelsden-Popper series of Ca<sub>n+1</sub>Ru<sub>n</sub>O<sub>3n+1</sub> aiming at tuning its structural transition by electric field was carried out. We found that the effect of electric field is minimal. We also carried out work on graphene, which was focused on the device fabrication and characterization on graphene flakes prepared by mechanical exfoliation from bulk graphite, and NbSe<sub>2</sub>, on the interplay between charge-density waves (CDWs) and superconductivity (SC) in atomically thin flakes again prepared by mechanical exfoliation from the bulk crystals. We observed strong magneto conductance fluctuations in few layer graphene near the charge neutral point, and novel electronic states at the “interface” between monolayer and bilayer graphene. For NbSe<sub>2</sub>, we found that the CDW was suppressed easily in the atomically thin NbSe<sub>2</sub> while SC survives at least down to two atomic layers.

## Publications

1. Xiangfan Xu, Zhuan Xu, Tijiang Liu, David Fobes, Zhiqiang Mao, and Ying Liu, "Band-dependent normal-state coherence in Sr<sub>2</sub>RuO<sub>4</sub>: Evidence from Nernst and thermopower measurements," **Phys. Rev. Lett.** 101, 057002 (2008).
2. J. A. Robinson, C. P. Puls, N. E. Staley, J. Stitt, M.A. Fanton, K. V. Emtsev, T. Seyller, Y. Liu, "Raman Topography and Strain Uniformity of Large-Area Epitaxial Graphene," **Nano Lett.** 9, 964-968 (2009).
3. C. P. Puls, N. E. Staley, Y. Liu, "Interface states and anomalous quantum oscillations in graphene hybrid structures," **Phys. Rev. B** 79, 235415 (2009).
4. Neal E. Staley, Linjun Li and Zhuan Xu, and Ying Liu, "Electric field effect on superconductivity in atomically thin flakes of NbSe<sub>2</sub>," **Phys. Rev. B** 80, 184505 (2009).
5. Yiqun A. Ying, Karl Nelson, Iosef G. Deac, Peter Schiffer, Peter Khalifah, Robert J. Cava, and Ying Liu, "Magneto electrical transport properties and possible quantum critical fluctuation in La<sub>4</sub>Ru<sub>6</sub>O<sub>19</sub>," **Phys. Rev. B** 80, 024303 (2009).
6. Y. A. Ying, Y. Xin, B. W. Clouser, E. Hao, N. E. Staley, R. J. Myers, L. F. Allard, D. Fobes, T. Liu, Z. Q. Mao, and Y. Liu, "Suppression of proximity effect and the enhancement of p-wave superconductivity in the Sr<sub>2</sub>RuO<sub>4</sub>-Ru system," **Phys. Rev. Lett.** 103, 247004 (2009).
7. Ying Liu, "Phase-sensitive measurement determination of odd-parity, spin-triplet superconductivity in Sr<sub>2</sub>RuO<sub>4</sub>," **New J. Phys.** 12, 075001 (2010).
8. Y. A. Ying, Y. Liu, T. He, and R. J. Cava, "Magnetotransport properties of BaRuO<sub>3</sub>: Observation of two scattering rates," **Phys. Rev. B** 84, 233104 (2011).

## Presentations

November 30, 2007. Department of Physics, University of Illinois in Urbana Champaign. "Pairing symmetries of the bulk and the 3-K phases of  $\text{Sr}_2\text{RuO}_4$ ."

December 11, 2007, *Workshop on Physics of  $\text{Sr}_2\text{RuO}_4$* , Kavli Institute for Theoretical Physics, University of California Santa Barbara. "Pairing symmetry of the 3-K phases of  $\text{Sr}_2\text{RuO}_4$ ."

January 15, 2009. Argonne National Laboratory, Chicago. "From Graphene to Single-Sheet Superconductors."

January 16, 2009. Northwestern University, Evanston. "Spin-triplet Superconductivity in  $\text{Sr}_2\text{RuO}_4$ ."

### **1. Transport properties of $\text{BaRuO}_3$**

We carried out transport measurements on  $\text{BaRuO}_3$  that features four crystalline structures for  $\text{BaRuO}_3$ , the nine-layered rhombohedral (9R), the four-layered hexagonal (4H), the six-layered hexagonal (6H), and the perovskite structure (3C). We measured only the 9R and 4H forms. It was found previously<sup>1</sup> that the 4H  $\text{BaRuO}_3$  shows a metallic resistivity down to the lowest temperatures. The 9R features a cross from metallic over to insulating behavior below 100K, which becomes flatten near 30K in the *c*-axis but not in the in-plane resistivity. The magnetic susceptibility of the 4H  $\text{BaRuO}_3$  was found to show a featureless, linearly increasing temperature dependence. However, the 9R  $\text{BaRuO}_3$  was found to possess a subtle change in slope around 100 - 150 K, slightly higher than the temperature below which the resistivity starts to cross over from an metallic to insulating behavior, and a more dramatic feature near 30 K.

We found that both forms of  $\text{BaRuO}_3$  show temperature dependent magnetoresistance. In particular, the behavior in the 9R  $\text{BaRuO}_3$  seems to correspond well with the behavior seen in resistivity and magnetic susceptibilities. More specifically, the magnetoresistance of the 9R  $\text{BaRuO}_3$  was found to be negative, growing rapidly below 30K. In comparison, the magnetoresistance of the 4H  $\text{BaRuO}_3$  was found to be positive, but becoming significant also below 30K. The Hall coefficient was found to be linear as a function of magnetic field up to 8 T and different in sign for 4H or 9R  $\text{BaRuO}_3$ , suggesting that there were no ferromagnetic instabilities in either form. Nevertheless, the magnetic fluctuation in 9R  $\text{BaRuO}_3$  is very different from that in 4H  $\text{BaRuO}_3$  at low temperatures, consistent with the magnetic susceptibility and resistivity results described above. Furthermore, the Hall coefficient was found to be nearly temperature independent for the 4H but not the 9R, suggesting that the 4H  $\text{BaRuO}_3$  is a good metal but the 9R is not.

The above observation is related to the structural features in these two forms of  $\text{BaRuO}_3$ . The 9R form consists of units of three  $\text{RuO}_6$  octahedra sharing faces in a partial chain, with a Ru-Ru distance of 2.529 Å, facilitating a metal-metal bonding. Each of these triple units of octahedra is connected to its neighbors along the hexagonal axis by perovskite-like corner sharing with the

nearly 180-degree Ru-O-Ru bonds. The stacking pattern repeats after 9 octahedra. The 4H form, on the other hand, consists of units of two octahedra sharing faces connected to each other by perovskite-type corner sharing. The stacking pattern along the hexagonal axis repeats after 4 octahedra. The distance between two Ru ions in the two face-sharing octahedra in the 4H BaRuO<sub>3</sub> is 2.537 Å. Both the 9R and the 4L forms of BaRuO<sub>3</sub> possess certain effective one dimensionality in the crystalline structure because of the presence of chains. In addition, both the 4H and the 9R BaRuO<sub>3</sub> feature a metal-metal bonding because of the very short Ru-Ru distance, leading possibly to the formation of local moments, resulting in the difference between the direct Ru-Ru and that of the oxygen mediated Ru-O-Ru interactions, and consequently, differences in electronic and magnetic properties.

The above result is published in Phys. Rev. B<sup>2</sup>.

## 2. Orbital ordering transition in ionic liquid-gated Ca<sub>3</sub>Ru<sub>2</sub>O<sub>7</sub> surface

While strontium ruthenates in the Roddelsden-Popper (R-P) series of Sr<sub>n+1</sub>Ru<sub>n</sub>O<sub>3n+1</sub> are all metals, the calcium ruthenates are more strongly correlated than their strontium ruthenate counterparts, featuring metallic as well as insulating behavior accompanied by magnetic and structural phase transitions. In particular, the bilayer calcium ruthenate, Ca<sub>3</sub>Ru<sub>2</sub>O<sub>7</sub>, features a band-dependent Mott metal-insulator transition (MIT) at 56 K, followed by a structural transition at 48 K as the temperature is lowered. Furthermore, a bulk spin valve behavior featuring colossal magnetoresistance was discovered, which was attributed to the existence of strongly spin-dependent resistive states in Ca<sub>3</sub>Ru<sub>2</sub>O<sub>7</sub>. The application of an in-plane field leads to a spin-reorientation as well as a resistive transition.

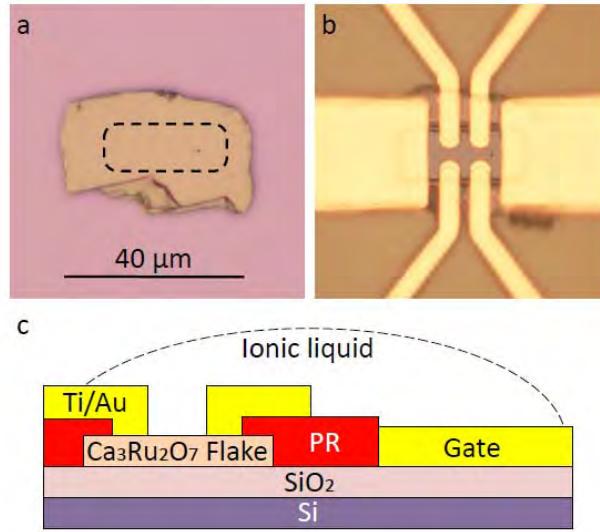


FIG. 1: a) Optical image of a Ca<sub>3</sub>Ru<sub>2</sub>O<sub>7</sub> flake supported by a Si/SiO<sub>2</sub> substrate. Dashed lines outline location of photoresist window to be patterned; b) Optical image of a device completed on the same flake with electrical contact made on the surface of the flake using a window through hard-baked photoresist; c) Schematic of the side-view of the device, including the coplanar ionic liquid gate (G) setup, with two of the six contacts acting as source (S) and drain (D).

We pursued the fabrication and measurements of ionic liquid gated Hall bar devices

prepared on very thin  $\text{Ca}_3\text{Ru}_2\text{O}_7$  flakes exfoliated from a bulk single crystals that were grown by a floating zone method (Fig. 1). We found that our process yielded two types of devices, Type A and Type B, where Type A devices are referred to those with their electrical transport properties dominated by c-axis transport and Type B devices by in-plane transport properties. Bulk physical phenomena, including a magnetic transition near 50 K, a structural and orbital ordering transition at a slightly lower temperature, as well as a highly unusual metallic state as the temperature is further lowered, were found in both types of devices (Fig. 2).

Interestingly, the Shubnikov-de Haas oscillations were found in Type A devices. These oscillations are consistent with those found in the bulk, suggesting that our Type A devices are of sufficient good quality. However, the same quantum oscillations were not observed in Type B devices. This is consistent with the idea that Type A devices are dominated by c-axis transport which is not sensitive to disorder on the surface while Type B devices are dominated by in-plane transport which most likely are subject to enhanced disorder on the flake surface. The most important result is that the ionic liquid gating of a Type B device did seem to lead to a shift in critical temperature of the structural and orbital ordering transition, suggesting that such a transition can be tuned by the electric field effect.

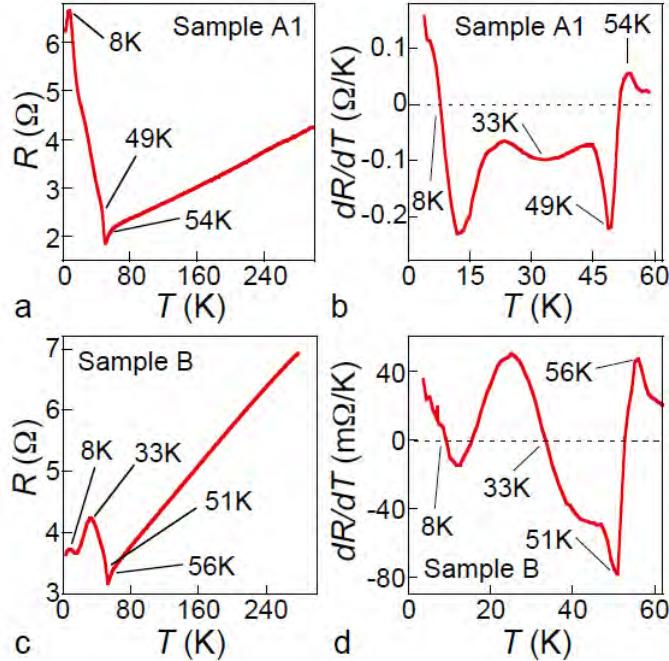


FIG. 2: a) Resistance  $R$  vs: temperature  $T$  in a  $\text{Ca}_3\text{Ru}_2\text{O}_7$  flake with low-T behavior dominated by c-axis transport; b)  $dR/dT$  vs:  $T$ , calculated numerically from (a), highlighting complex transition behavior at low temperatures; c)  $R$  vs:  $T$  in a  $\text{Ca}_3\text{Ru}_2\text{O}_7$  flake with low-T behavior dominated by in-plane transport; d)  $dR/dT$  vs:  $T$ , calculated numerically from (b).

## References

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<sup>1</sup> J. T. Rijssenbeek, R. Jin, Y. Zadorozhny, Y. Liu, B. Batlogg, and R. J. Cava, “The Electrical and Magnetic Properties of the Two Crystallographic Forms of  $\text{BaRuO}_3$ ,” **Phys. Rev. B** 59 4561-4564 (1999).

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<sup>2</sup> Y. A. Ying, Y. Liu, T. He, and R. J. Cava, “Magnetotransport properties of BaRuO<sub>3</sub>: Observation of two scattering rate s,” **Phys. Rev. B** 84, 233104 (2011).